

AD-A119 772

HOWARD UNIV WASHINGTON DC DEPT OF CHEMISTRY
PHOTODISSOCIATION DYNAMICS IN A PULSED MOLECULAR BEAM. (U)
SEP 82 R LU, J B HALPERN, W M JACKSON

F/G 20/5

N00014-80-C-0305

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REPORT DOCUMENTATION PAGE		
1. REPORT NUMBER ADR-TR-13-ONR	2. GOVT ACCESSION NO. AD-A119772	3. REPORT TYPE AND PERIODICITY
4. TITLE (and Subtitle) PHOTODISSOCIATION DYNAMICS IN A PULSED MOLECULAR BEAM	5. TYPE OF REPORT & PERIOD COVERED TECHNICAL REPORT 13	
7. AUTHOR(s) Richang Lu, J. B. Halpern and W. M. Jackson	6. PERFORMING ORG. REPORT NUMBER	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Laser Chemistry Division Department of Chemistry Howard University Washington, D. C. 20059	8. CONTRACT OR GRANT NUMBER(s) N00014-80-C-0305	
11. CONTROLLING OFFICE NAME AND ADDRESS Office of Naval Research Department of the Navy Arlington, VA22217	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS NR-051-733	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	12. REPORT DATE September 16, 1982	
	13. NUMBER OF PAGES 21	
	15. SECURITY CLASS. (of this report) UNCLASSIFIED	
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; reproduction is permitted for any purpose of the United States Government; distribution is unlimited.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report) Distribution of this document is unlimited.		
19. SUPPLEMENTARY NOTES Prepared for presentation at the Conference on Lasers as Reactants and Probes in Chemistry held at Howard University on May 12-14, 1982. To be published in the proceedings of the meeting, 1983.		
20. KEY WORDS (Continue on reverse side if necessary and identify by block number) Photodissociation, excimer laser, ClCN, C₂N₂, cyanogen chloride, cyanogen, seeded beams		
21. ABSTRACT (Continue on reverse side if necessary and identify by block number) The ArF laser photodissociation of C₂N₂, ClCN and BrCN have been studied in a pulsed beam. Comparisons are made between these results and the radical product distributions measured in an effusive beam. Comparisons are also made between the effects of cooling by expansion in argon and expansion in methanol. The latter is known to more effectively cool vibrational modes in the parent molecule.		

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Contract N00014-80-C-0305

Task No. NR 051-733

TECHNICAL REPORT NO. 13

Photodissociation Dynamics in a Pulsed Molecular Beam

by

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Prepared for Publication

in the

Proceedings of the

Conference on Lasers as Reactants

and Probes in Chemistry

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September 17, 1982

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**Photodissociation Dynamics in a Pulsed
Molecular Beam**

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ABSTRACT

The Arf laser photodissociation of C_2H_2 , $ClCN$ and $BrCN$ have been studied in a pulsed beam. comparisons are made between these results and the radical product distributions measured in an effusive beam. Comparisons are also made between the effects of cooling by expansion in argon and expansion in methane. The latter is known to more effectively cool vibrational modes in the parent molecule.

Introduction

The nascent quantum state distributions of the fragments yield important information about the details of the photodissociation process. The presence of large amounts of rotational energy in the fragments indicates that substantial geometrical changes have occurred when the system went from the ground to the excited state. Of course when photodissociation is studied under bulb conditions at room temperature the molecules exist with a distribution of rotational and vibrational energies. The bending vibrations of the molecule have very low fundamental frequencies so that a substantial population is expected in the first few levels at 300 K. It is, therefore, not completely clear how much of the rotational energy observed in the fragments is due to this vibrational excitation. Further in the case of transitions between a linear ground state and a linear excited state our earlier work (1) has suggested that the observed angular momentum of the fragments is due to the original angular momentum present in the parent molecule. To sort out such effects it is important to experimentally vary the original amount of angular momentum in the parent molecule and determine how this affects the angular momentum observed in the fragments.

Free jet expansion in seeded molecular beams is known to cool the internal degrees of the diluent molecule. Pulsing these molecular beams reduces the required vacuum pump overhead and is consistent with the use of a pulsed tunable dye laser to determine the quantum state distribution. With this technique internal rotational and vibrational temperatures as low as 1 K and 50 K respectively have been obtained depending upon the

experimental conditions that are chosen. Thus the use of pulsed molecular beams allows one to vary the amount of vibrational and rotational energy that is present in the molecule before photoexcitation.

For several years various workers in our laboratory have been involved in measuring the nascent quantum state distributions of fragments produced in the photolysis of linear cyanide containing molecules (1-5). There are many reasons why these systems have been chosen for systematic study. First many of the theoretical calculations on photodissociation of simple molecules have been done for the triatomics HCN or ICN (6-10). Second, HCN, ClCN, BrCN and ICN provide a simple homologous series of triatomic compounds which along with C_2N_2 , a tetratomic compound, with quasi-diatomic characteristics are all commercially available. Thirdly, CN is an ideal molecule for laser induced fluorescence detection. The lifetime of the $B^2\Sigma^+$ state is short enough, about 60ns [11], that the fluorescent photocurrent at the detection photomultiplier will be high, but long enough that scattered laser light may be eliminated by using a gated boxcar analyzer. The excitation spectrum is simple, consisting of a single P and R branch for each vibrational band, and the separation between vibrational levels is small enough so that a single dye can be used to scan several vibrational bands, but large enough so that the spectra are not completely overlapped. Moreover, we have shown how LIF can also be used to measure the quantum state distribution of any CN radicals produced in the low lying $A^2\Pi_1$ electronic state (12).

Recently we reported on the photolysis of C_2N_2 (1), ClCN and BrCN (3) at 193 nm by an ArF laser in an effusive beam. The results may be

summarized as follows: In the case of C_2N_2 , there are two ground state CN fragments produced as a result of predissociation through the ground state vibrational continuum of the $X^1\Sigma^+$ state of C_2N_2 . Roughly 35% of the CN fragments are produced in the $v''=1$ state. The rotational distribution of both the $v''=0$ and $v''=1$ fragments can be described by a 900K Boltzmann distribution. This distribution can be reproduced by assuming that the excited state of the molecule is also linear and that each of the CN fragments carries away half of the rotational angular momentum of the parent, which itself follows a 300K Boltzmann distribution. ClCN and BrCN, on the other hand, distribute about half of the available energy into rotation. The total amount of this excess energy ranges from about 18,000 cm^{-1} for ClCN and about 22,000 cm^{-1} for BrCN. The rotational distributions of CN fragments are non-Boltzmann for all vibrational levels, being rotationally inverted with respect to thermal distributions. For the case of ClCN about 30% of the fragments are produced in $v''=1$, 19% in $v''=2$ and a smattering of population is observed in $v''=3$. For BrCN most of the population is found in $v''=0$ while only a small amount of $v''=1$ fragments are seen. We have been able to parameterize the rotational distributions with two parameters, in a model which will be discussed below.

By expanding the parent molecule in a seeded supersonic molecular beam it is possible to cool the quantum state distribution of the parent molecules. This will give us more information about the photodissociation process, since we can determine how the angular momentum in the original parent molecule affects the angular momentum of the fragments.

Experimental

The experimental apparatus which has been previously described, (1,5) consists of dye and excimer laser beams counter-propagating and crossing a molecular beam in the experimental cell. Induced fluorescence is observed through a filtered and apertured photomultiplier placed at right angles to the lasers. In the experiments described here a pulsed beam source is used. The pulsed beam is formed using a pulsed valve with a 0.2mm i.d. orifice plate. The valve opens with a 500 μ sec rise and fall time and is on for 3 μ sec. The lasers were triggered so that light shined on the molecular beam 350 μ sec from the initial rise of the pulsed molecular beam. The background pressure in the reaction cell rose to about 5×10^{-5} torr when the pulsed valve was running at 10 Hz. In some experiments a big hole, 0.8mm i.d. and a lower frequency, 1 Hz, were used to enhance the cooling effect.

Figure 1 shows sample excitation spectra of CN fragments produced from the photodissociation of both an effusive and a pulsed beam of C_2N_2 . The pulsed beams use Argon or CH_4 as the carrier gas. It is clear that there is a marked shift of peaks in both the P and R branch. This indicates that the radicals have a lower rotational temperature. Detailed rotational state analysis confirms that the rotational distributions have been cooled.

The rotational cooling that is observed in the CN fragments from C_2N_2 is however much less than one would predict based upon our earlier model

(1). This model suggests that the temperature observed in the CN fragment should be approximately three times the original rotational temperature of the C_2N_2 in its ground electronic state. Table 1 shows that under a variety of conditions with both CH_4 and Ar carrier gases the rotational temperature of the CN product was never below 540K which according to our original model would correspond to a parent rotational temperature 180K. However, the rotational temperature of C_2N_2 in some of these pulsed beam experiments has been much lower than this.

To test whether rotational cooling is actually occurring in the pulsed molecular beam, an experiment was performed where CN was deliberately formed in the earlier part of the beam before the supersonic expansion was complete. The result of this experiment is shown in Figure 2. The CN radical has been substantially cooled since most of the population occurs in the first few J levels. This indicates that a great deal of rotational cooling occurs in our pulsed beam system as one would expect. The "temperature" one would calculate from this experiment is <10K. Therefore, the results reported in Table 1 cannot be explained in terms of incomplete expansion and poor rotational cooling of the $X^1\Sigma$ state of C_2N_2 .

As a sidelight it is interesting to note that there is a small P_{00} bandhead observed in the spectra. This further confirms repeated observations by us in bulb experiments that the rate of rotational cooling for the upper J levels is less than it is for the lower levels.

The results of the cooling experiment have forced us to conclude that the agreement between our earlier model and experiment was fortuitous. The predictions of the model do not agree with the present observations.

What then can be used to explain the present observations? Freed et. al. (9) have suggested that an additional source of rotational angular momentum of the products for a linear to linear transition is the conversion of the angular momentum tied up in the bending vibrations. This would not disagree with the present observations that rotational cooling of the parent in the supersonic expansion does not result in large amounts of cooling in the product.

This does not, however, explain all of the results. The optical transition that we are studying is a ${}^1\Sigma_g$ to ${}^1\Delta_u$ transition. Such a transition is optically forbidden since $\Delta\Lambda$ is +2 which violates the orbital selection rule. The absorption coefficient for the transition is of the order of 10^{-19} cm² which though small does not correspond to an optically forbidden transition. The transition can be made optically allowed by mixing in the symmetry of the bending modes with the electronic symmetry. This is illustrated in Figure 3 where an energy level diagram has been drawn for an optically allowed transition from the (00000) and (00010) levels of the $X{}^1\Sigma_g$ state of C_2N_2 to the upper levels of $B{}^1\Delta_u$ of the molecule. By mixing in the vibrational symmetry of the bending modes with the electronic symmetry the transition is now allowed. The small observed absorption coefficient is then the result of poor Franck-Condon factors for the transition.

So far all of the results that have been discussed do not disagree with the Freed model. Two observations, however, are in apparent disagreement. First in an effusive beam at 300 K the rotational distribution of both the $v''=1$ and the $v''=0$ levels of the CN radicals are the same.

of the parent state. The results indicate that the relative vibrational population of the fragments increases with increasing expansion ratio. Further, it is found that the carrier gas is the carrier gas that it is when Argon is the carrier gas. Previous work (13) has shown that principal difference between these two should be the efficiency of vibrational cooling in the beam. Thus it would seem that the change in vibrational population of the fragment is associated with the vibrational population of the C_2N_2 parent. The bending vibrations of C_2N_2 are 233.1 cm^{-1} and 506 cm^{-1} for ν_3 and ν_4 frequencies respectively so that significant amount of bending states will be populated at room temperature. The $v''=1$ level of CN may result from the 00001 level of C_2N_2 while the $v''=0$ level may result from the 00000 level of the molecule. Even though the fragments arise from different vibrational levels of the molecule the observed rotational distributions are the same in contradiction to Freed's model. Further, his model predicts that large oscillations should be observed in the rotational distribution of the fragments that result from a parent where the bending mode is excited. Within experimental error no such quantum beats are observed. Since tetratomic molecules are more complicated there may be other effects that wash out these quantum beats.

The spectra in Figure 3 show that there is almost no observed change in the rotational distribution of CN fragments produced in the photolysis of CICH in a pulsed molecular beam. The only noticeable change is a decrease in the vibrational population of $v''=1$. This decrease is

8

within the experimental error. For BrCN there is a slight change in the rotational distribution of the CN fragments as can be seen in Figure 4. This is also within the experimental error. These results confirm the original observations (5) that the transition that was excited in ClCN and BrCN by the 193 nm laser is a linear to bent transition. Careful consideration shows that only highly excited levels of the bending mode can be reached in such a process. The reason for this can be seen in Figure 5 where the potential energy associated with bending is shown as a function of angle. Because this potential must be symmetric about the linear geometry the excited state will show a local minimum at 180° . The repulsive walls shown at zero and 360° just attest to the fact that the atoms at the ends of the triatomic molecule cannot interpenetrate. In general, there will be a difference in energy between the local maximum and the vibrational level in the excited state reached by absorption. If this difference is small, the linear geometry will be a stationary point of the vibrational motion of the molecule in the excited state and the Franck-Condon overlap will be large. If this difference is large, the overlap will be vanishing small and there will be very little absorption. This would explain the weak absorption coefficient, 10^{-19}cm^2 , of these compounds in the first continuum [14].

Large differences in energy between the local maximum and the vibrational level that is accessed suggest that the bending vibrational motion of the excited molecule will be fast. This is in agreement with the observation of large amounts of rotational energy being found in the fragment. If the bending vibrational motion was slow relative to

dissociation then the rotational energy that appears in the fragment would be appreciably reduced.

Earlier we were able to parameterize the effusive rotational distributions by choosing the maximum rotational quantum number of the fragment L , as one of the parameters, and then plotting the distribution $P(L-J)$. This turned out to be a Boltzmann like distribution, describable by a single temperature. We speculated that the distribution was a remnant of the rotational distribution of the parent, with the angular momentum of the CN being simply related to that of the ClCN. If this were true one would expect that an observable change in the CN distribution would occur as the parent was cooled, the present results indicate that this has not happened. This may be because at 300K the contribution of the parent's rotation to the rotational energy of the fragments is small relative to the contribution from the vibrational motion of excited molecule. Thus, a change in the parent's rotational energy by cooling does not affect the fragment energy.

Conclusions

Pulsed beam studies on the photolysis of C_2N_2 at 193 nm have yielded unexpected results that the reduction of the rotational temperature of the parent does not correspond to an observed reduction in the rotational temperature of the CN fragment. This is in contradiction to our conclusion based on earlier result in an effusive beam. Compare our experimental results with the theoretical predictions of Freed et al. While some of the observed rotational energy in CN is related to the original vibrational

motion of C_2N_2 other detailed predictions do not agree with the experimental observations. In particular the rotational temperatures of the $v''=0$ and the $v''=1$ levels of CN are the same for an effusive beam and for a pulsed molecular beam. Arguments have been presented that suggest that these two fragments come from different bending vibrational levels of the ground states of C_2N_2 . The Freed model would then suggest that the rotational temperatures of the two fragments would have to be different.

The present results may be rationalized in the following manner. The photodissociation process that we are investigating in C_2N_2 is really a predissociation through the vibrational continuum of the ground state. The rotational distribution will be determined by the population of the bending modes in the excited state just prior to its crossing over into the vibrational continuum of the ground state. The geometry of this state is the determining factor for the rotational distribution of the fragment. Since the rotational distribution is the same for the $v''=1$ and $v''=0$ levels, it suggests that only a certain geometrical configuration has a high probability for crossing over to the vibrational continuum of the ground state.

If the ideas that have been presented for CN from C_2N_2 are correct, then one would expect that changing the wavelength for dissociation would not change the observed rotational distribution of CN. The reason is that this rotational distribution is fixed by "Franck-Condon" envelopes between the B^1_u state prior to cross over and the vibrational continuum of the ground state. Work is in progress to shift the wavelength of the dissociating laser to determine whether this idea is correct.

The pulsed beam studies on ClCN and BrCN are in agreement with the idea that the rotational distribution that is observed is determined by the Franck-Condon factors or overlap between the ground vibrational states of the halogen containing compound and the excited bent state of these compounds. Slightly different configurations in the excited states are apparently associated with a CN product in a different vibrational states. The justification of this statement is that the rotational distributions of the $v''=0, 1$ and 2 for CN are different for each of these vibrational states [5]. Note, however, that BrCN is only produced in one vibrational state. In the effusive beam studies we ascribed this behavior to the fact that a part of the upper potential surface that is access upon photo-excitation is much flatter for BrCN than it is for ClCN. Hence, the excited molecule has much less vibrational excitation in BrCN so much less shown up in the CN fragment. The pulsed beam studies do not contradict that observation, but neither do they lend any credence to it. Further studies at different wavelengths should show a different amount of vibrational excitation in the CN fragments for these two compounds.

Acknowledgement

William M. Jackson would like to acknowledge the support of the Office of Naval Research for his summer salary and for support for some of the equipment on this grant. Dr. J. B. Halpern acknowledges the support of NASA under grant NAG-517 for his summer salary and Dr. Richard Lu acknowledges the support of the Department of Energy under grant 9000-3034 for his support as a visiting scholar along with partial support for the molecular beam apparatus that was used on this grant.

Figure Captions

- Figure 1. LIF excitation spectra of $\text{CN}(X^2\Sigma^+)$ produced in the photodissociation of C_2N_2 at 193 nm (a) in effusive beam and (b) in pulsed supersonic beam.
- Figure 2. Transitions allowed by mixing electronic and bending vibrational symmetries of C_2N_2 .
- Figure 3. LIF excitation spectrum of $\text{CN}(X^2\Sigma^+)$ produced and cooled in the upstream of pulsed beam. Only a few low J levels are strongly distributed. The figure shows R-branch of $\Delta v=0$ sequence.
- Figure 4. Rotational distribution of $\text{CN}(X^1\Sigma^+)$ from ClCN photolysis at 193 nm. Only $v''=0$ level is shown, and the highest rotation quantum number, J max, is 73.
- Figure 5. Rotational distribution of $\text{CN}(X^1\Sigma^+)$ from BrCN photolysis at 193 nm. The highest rotational quantum number. J max is 77.
- Figure 6. Schematic diagram of a linear to bent transition. θ is the bending angle. See text for details.

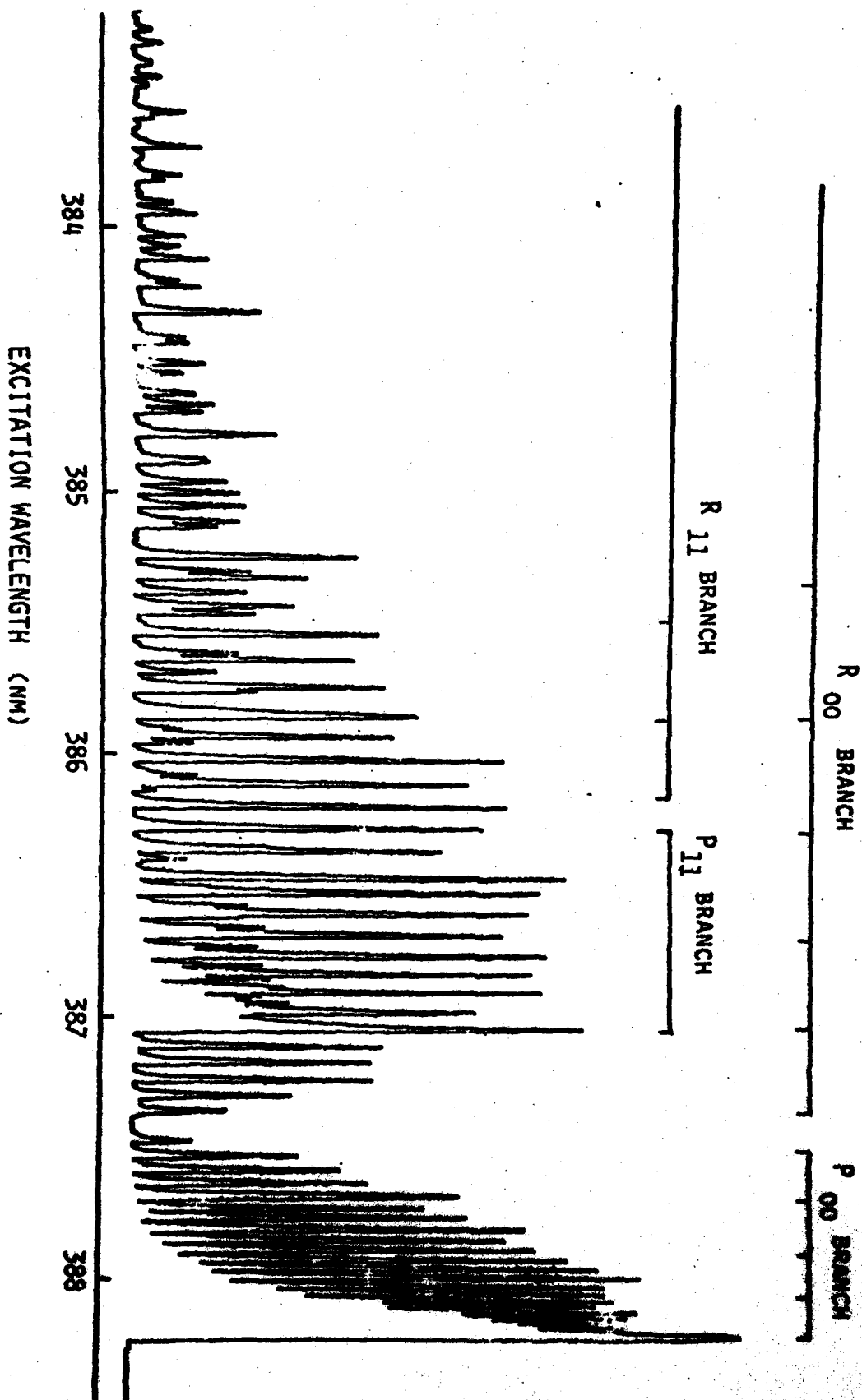
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BEAM	GAS		T_R (K)	$N_V''=1/N_V''=0$
	CARRIER	P (torr)		
EFFUSIVE PULSED " " " "	Ar CH ₄ CH ₄	<0.1	900	0.35
		410	800	0.22
		560	620	0.12
		730	540	0.11

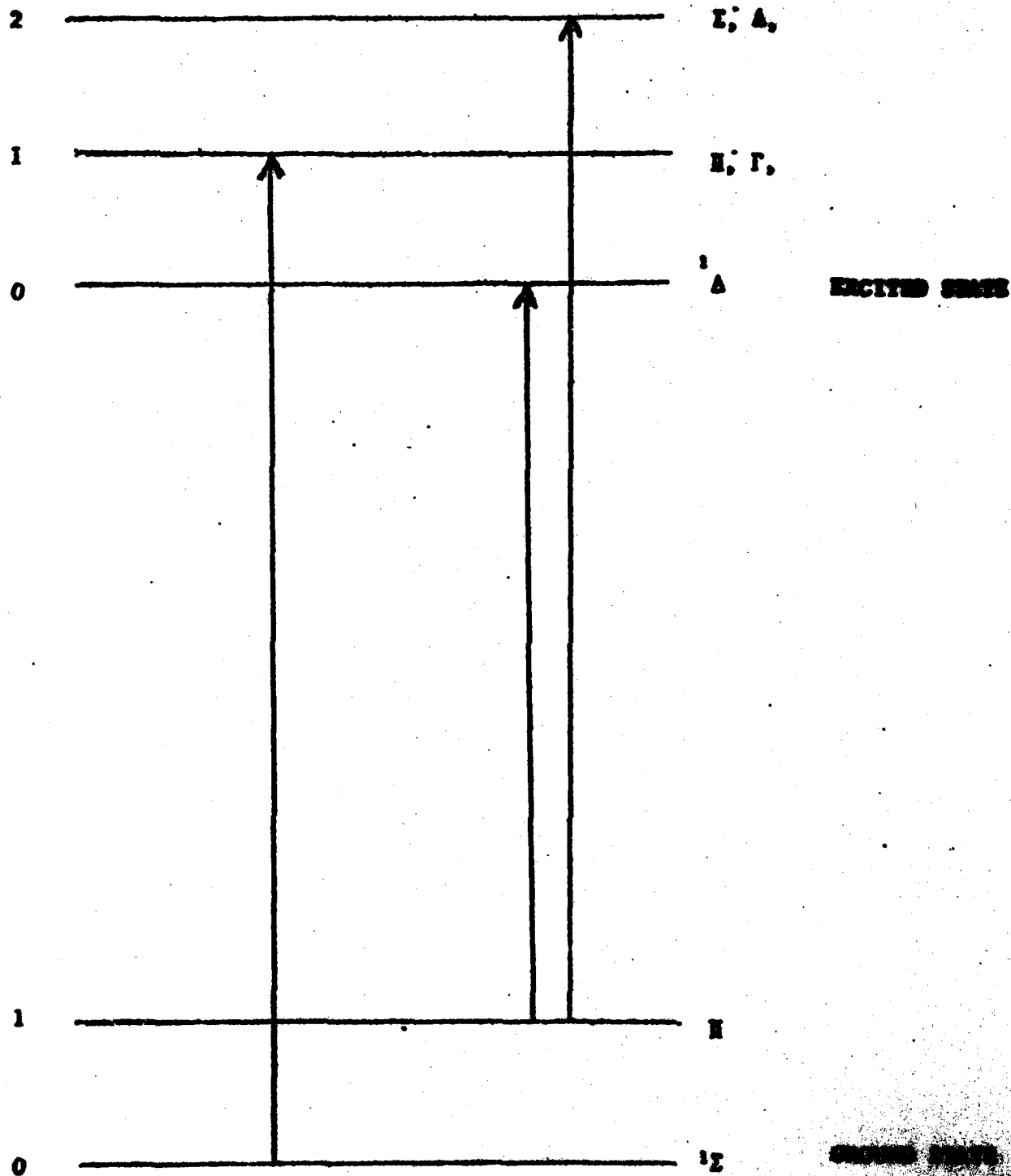
Table 1

C_2N_2 PHOTOLYSIS AT 193 NM
CN PRODUCT LASER EXCITATION SPECTRUM



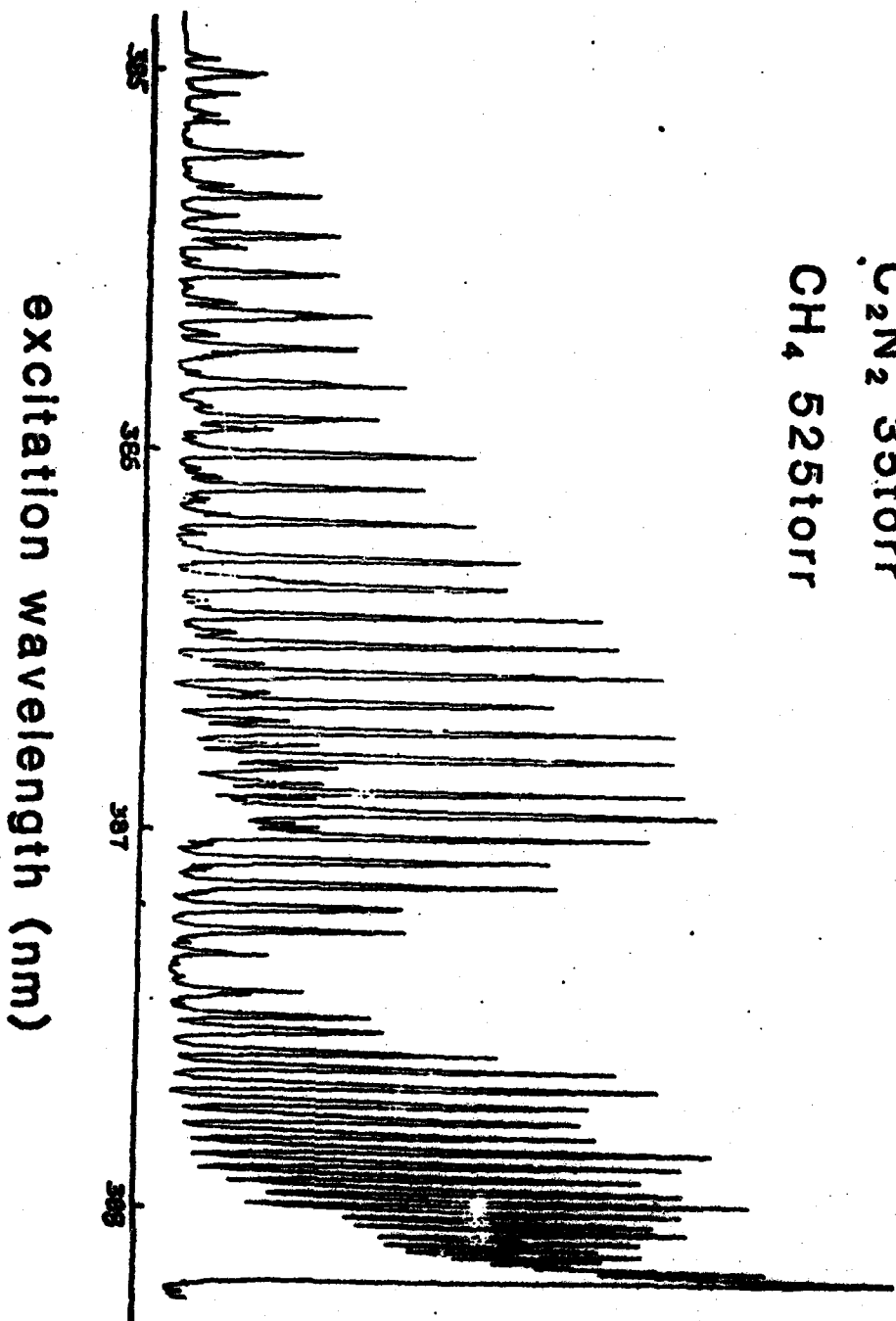
**QUANTA IN
BENDING MODES**

**OVERALL
SYMMETRY**

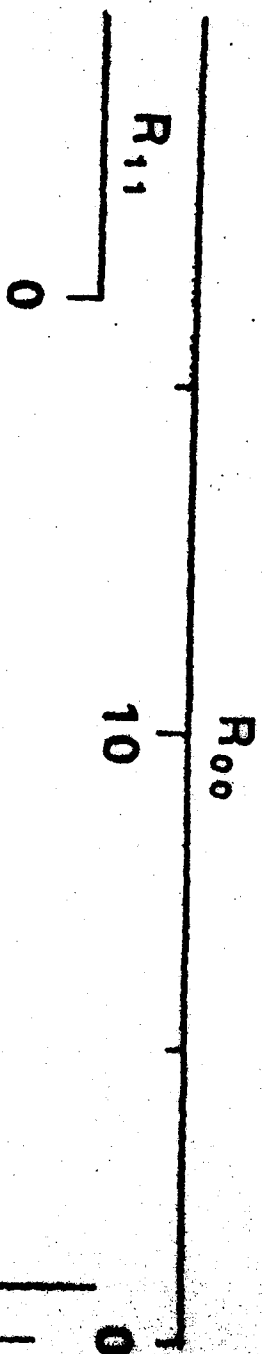


C_2N_2 PHOTOLYSIS IN A PULSED BEAM AT 103nm

C_2N_2 35torr
 CH_4 525torr



R BRANCH OF CN COOLED IN PULSED BEAM

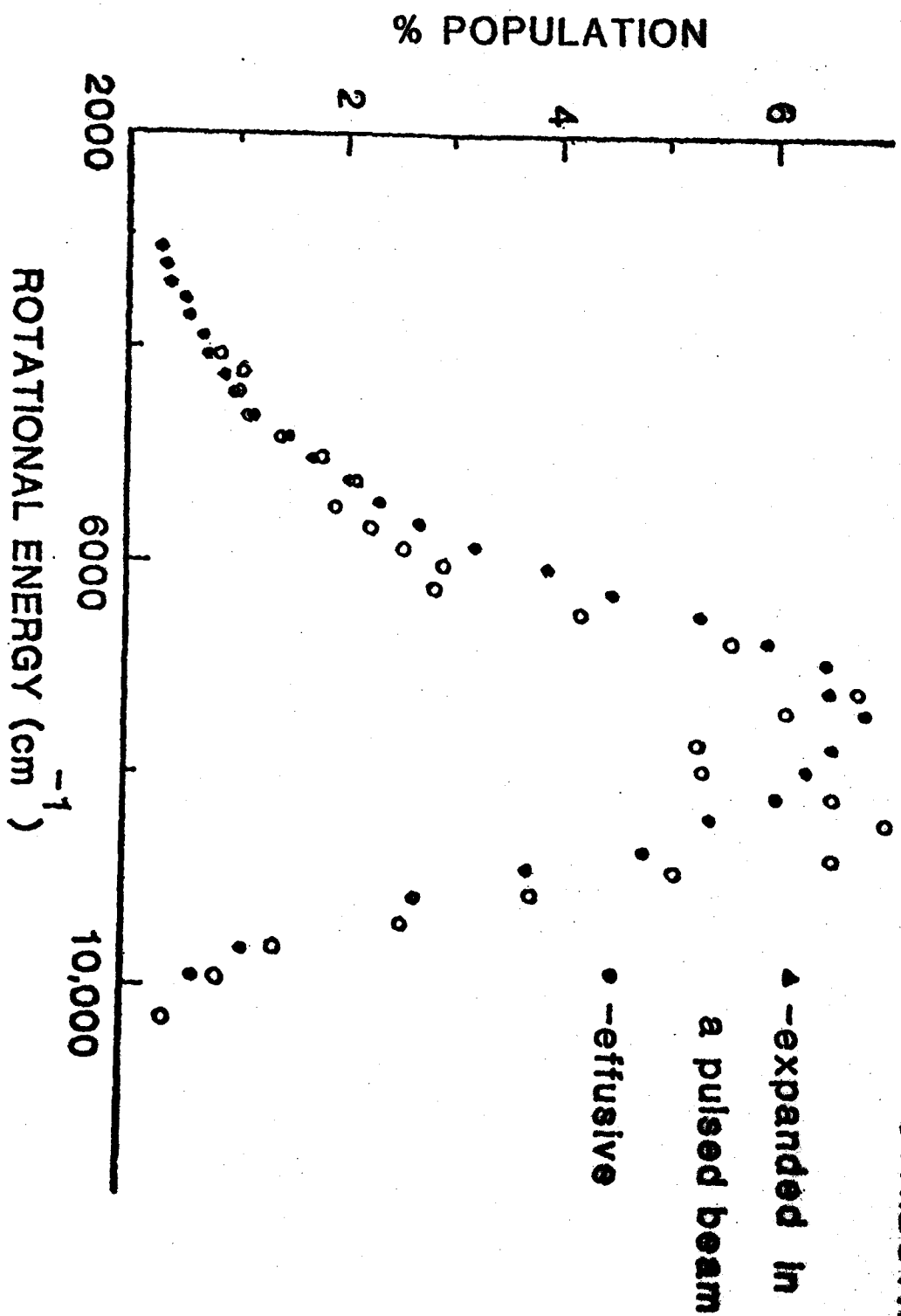


730 torr Ar
15 torr C₂N₂

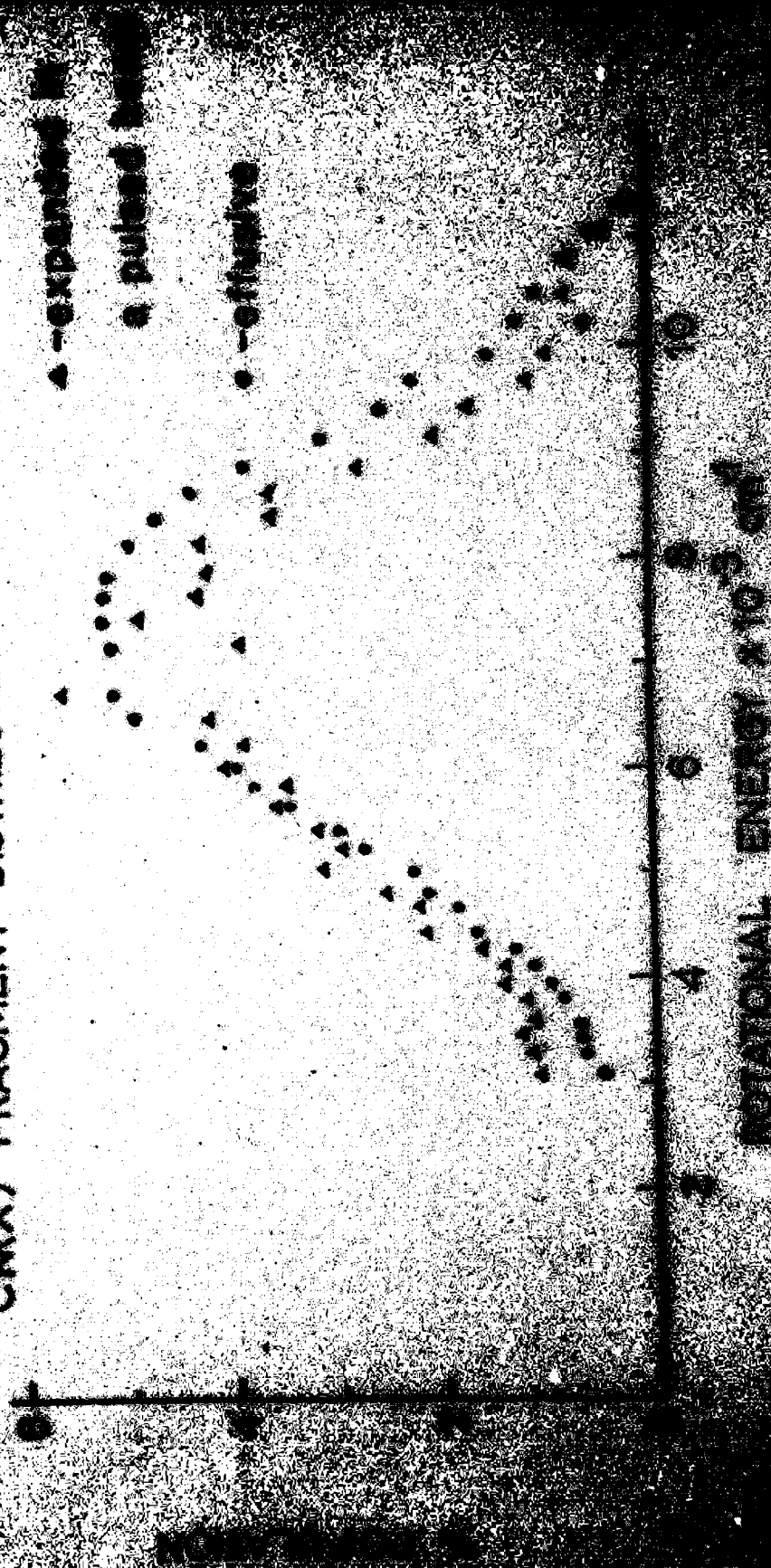
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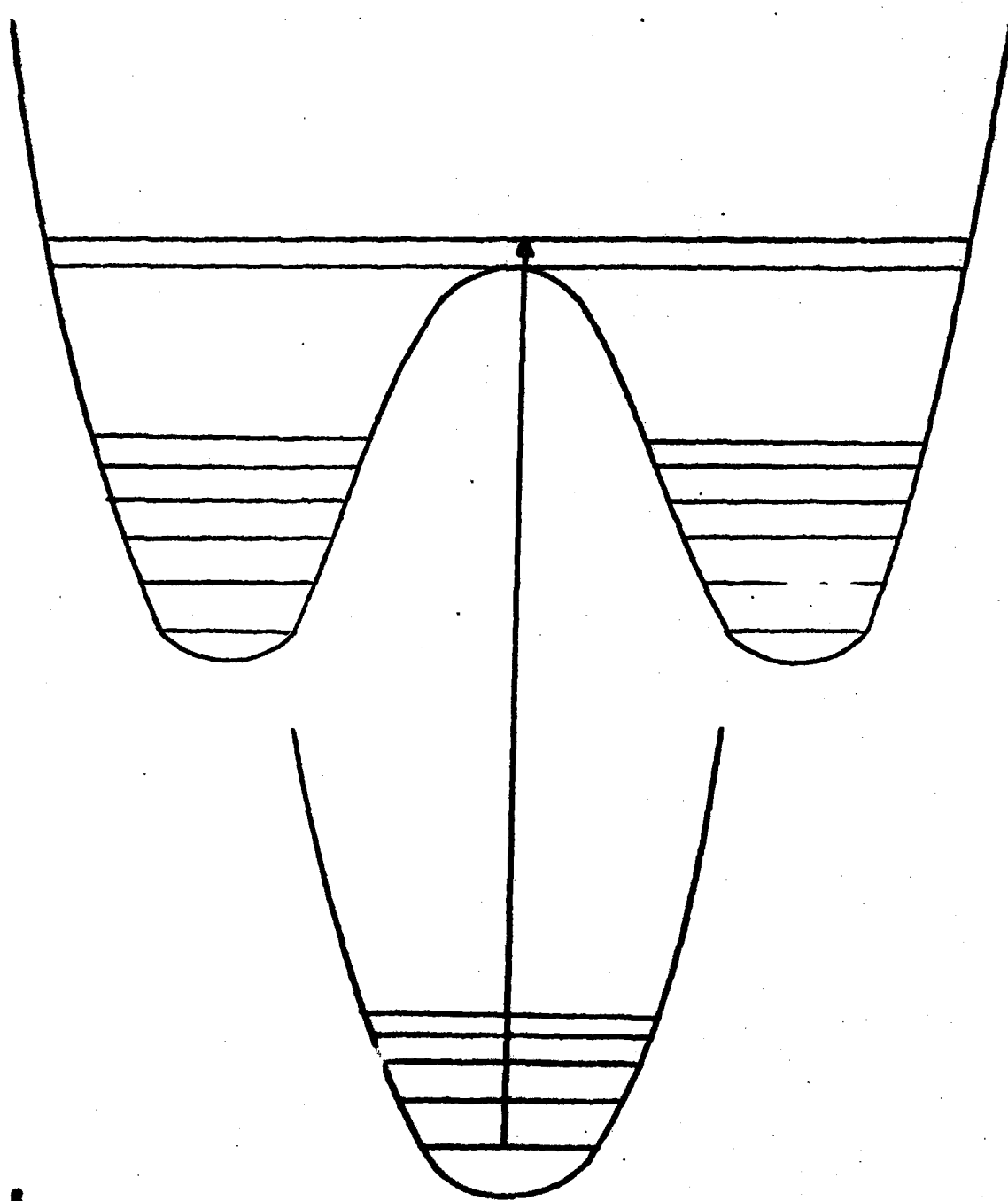
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CLCN PHOTOLYSIS AT 193nm CN(X) FRAGMENT DISTRIBUTION



BrCN PHOTOLYSIS AT 193 nm CN(X) FRAGMENT DISTRIBUTION





0

180

360

Θ = Bending Angle

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